(12) UK Patent Application (19) GB (11) 2 290 296 (13) A

(43) Date of A Publication 20.12.1995

(21) (22) (30)	Priority Data (32) 13.06.1994 (33) GB	FICE (52) .E CT UAL	INT CL ⁶ C08K 5/3412 5/3415 5/3417 5/3432 UK CL (Edition N) C3K KSA K210 K211 K272 K274 K282 K291 K293 K294 EL C3W W100 W216 W217 W218 W219 W308 W314 W317 W322
(71)	Applicant(s) Sandoz Ltd (Incorporated in Switzerland)	3 1988	Documents Cited GB 1224721 A EP 0574804 A2 EP 0410514 A1 EP 0373900 A2 EP 0337814 A2 EP 0295103 A2 US 5362808 A US 4311836 A
	35 Lichtstrasse, CH-4002 Basle, Switzerland		WPI Abstract Accession No:71-04536S/03 & DE1935239B
(72)	Inventor(s) Peter Staniek Klaus Stoll	(58)	Field of Search UK CL (Edition N) C3K KSA KXX INT CL ⁶ C08K 5/3412 5/3415 5/3417 5/3432 5/3437 ONLINE: PATENTS
(74)	Agent and/or Address for Service B A Yorke & Co Coomb House, 7 St John's Road, Isleworth, Middlesex, TW7 6NH, United Kingdom	i i	

(54) Imide nucleating agents for polymers

(57) A polymeric composition comprises at least one polymeric material and at least one nucleating agent selected from a group consisting of specified imide compounds. More particularly a process is provided to improve the degree of crystallinity, the crystallization onset temperature and/or the mechanical properties of polymeric materials. The use of the specified imides as nucleating agents for polymeric materials as well as to compositions, such as masterbatches and liquid concentrates, comprising said imide compounds is described.

-1-		(WPAT)	
PŅ	-	GB2290296-A	95.12.20
		DE19520262-A1	95.12.14
		FR2721036-A1	95.12.15
	•	JP08012809-A	96.01.16
DNIA	_	T00012000	

NUCLEATING AGENTS

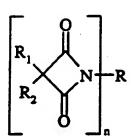
The invention relates to a polymeric composition comprising at least one polymeric material and at least one nucleating agent selected from the group consisting of compounds of formulae I to VIII.

More particularly the invention relates to a process to enhance the degree of crystallinity, the crystallization onset temperature, and the mechanical properties of polymeric materials. The invention relates further to the use of special imide compounds according to formulae I to VIII as nucleating agents for polymeric materials as well as to compositions, e. g. masterbatches and liquid concentrates comprising said imide compounds.

The physical properties which are improved by these nucleating agents (hereinafter called NA), are for example, crystallization temperature and/or mechanical properties like tensile strength or elongation at break.

According to the invention, there is provided a polymeric composition comprising a polymeric material and a nucleating agent of formulae I to VIII

(II)
$$\begin{bmatrix} R_1 & R_1 & R_2 & R_2$$



(III)

$$\begin{bmatrix} R_1 & R_1 & O \\ R_2 & & N \\ R_2 & & N \end{bmatrix}_n$$

$$\begin{bmatrix} R_5 & X & O \\ R_1 & R_5 & X & O \\ R_2 & R_5 & O \end{bmatrix}_{\mathbb{R}}$$

(VIII)
$$R_6$$

in which

R if n=1, is

hydrogen, C_{1-24} alkyl (linear or branched), C_{5-12} cycloalkyl, C_{6-24} aryl, C_{4-24} heteroaryl, C_{7-24} aralkyl, C_{7-24} alkaryl, $-C_{1-24}$ alkylen- R_7 ;

particularly hydrogen, C_{1-18} alkyl (linear or branched), C_{5-8} cycloalkyl, C_{6-18} aryl, C_{4-18} heteroaryl, C_{7-18} aralkyl, C_{7-18} alkaryl, $-C_{1-18}$ alkylen- R_7 ;

preferably hydrogen, C_{1-12} alkyl (linear or branched), C_{5-8} cycloalkyl, C_{6-18} aryl, C_{4-12} heteroaryl, C_{7-18} aralkyl, C_{7-18} alkaryl, $-C_{1-18}$ alkylen- $-R_7$;

more preferably hydrogen, C_{1-12} alkyl (linear or branched), cyclohexyl, phenyl, biphenyl, C_{7-12} aralkyl, C_{7-12} alkaryl;

if n>1

 $C_{1.24}$ alkylen (linear or branched), $C_{5.12}$ cycloalkylen, $C_{6.24}$ arylen, $C_{4.24}$ heteroarylen, $C_{7.24}$ aralkylen, $C_{7.24}$ alkarylen;

particularly C_{1-18} alkylen (linear or branched), C_{5-8} cycloalkylen, C_{6-18} arylen, C_{4-18} heteroarylen, C_{7-18} aralkylen, C_{7-18} alkarylen;

preferably C_{1-12} alkylen (linear or branched), C_{5-8} cycloalkylen, C_{6-12} arylen, C_{4-12} heteroarylen, C_{7-18} aralkylen, C_{7-18} alkarylen;

more preferably C_{1-2} alkylen (linear or branched), cyclohexylen, phenylen, biphenylen, C_{7-12} aralkylen, C_{7-12} alkarylen;

R₁, R₂, R₃ are independently selected from hydrogen, C₁₋₂₄alkyl (linear or branched), C₅₋₁₂cycloalkyl, C₆₋₂₄aryl, C₄₋₂₄heteroaryl, C₇₋₂₄aralkyl, C₇₋₂₄alkaryl, -NHR₄;

particularly hydrogen, C₁₋₁₈alkyl (linear or branched), C₅₋₈cycloalkyl, C₆₋₁₈aryl, C₄₋₁₈heteroaryl, C₇₋₁₈aralkyl, C₇₋₁₈alkaryl, -NHR₄;

preferably hydrogen, C_{1-12} alkyl (linear or branched), C_{5-8} cycloalkyl, C_{6-18} aryl, C_{7-18} aralkyl, C_{7-18} alkaryl;

more preferably hydrogen, C₁₋₁₂alkyl (linear or branched), cyclohexyl, phenyl, biphenyl, C₇₋₁₂aralkyl, C₇₋₁₂alkaryl;

R₄ hydrogen, C₁₋₂₄alkyl (linear or branched), C₅₋₁₂cycloalkyl, C₆₋₂₄aryl, C₄₋₂₄heteroaryl, C₇₋₂₄aralkyl, C₇₋₂₄alkaryl, -C₁₋₂₄alkylen-R₇;

particularly hydrogen, $C_{1.18}$ alkyl (linear or branched), $C_{5.8}$ cycloalkyl, $C_{6.18}$ aryl, $C_{4.18}$ heteroaryl, $C_{7.18}$ aralkyl, $C_{7.18}$ alkaryl, $-C_{1.18}$ alkylen- R_7 ;

preferably hydrogen, $C_{1.1}$ alkyl (linear or branched), $C_{5.8}$ cycloalkyl, $C_{6.18}$ aryl, $C_{7.18}$ aralkyl, $C_{7.18}$ alkaryl;

more preferably hydrogen, $C_{1.8}$ alkyl (linear or branched), cyclohexyl, phenyl, $C_{7.12}$ aralkyl, $C_{7.12}$ alkaryl;

- R₅ hydrogen, C_{1.24}alkyl (linear or branched), -CH₂-OH; particularly hydrogen, methyl;
- R₆ -OH, -NHR₄;

 R_7

- X -CH₂-, >CH-C₁₋₄alkyl- oder -O-; particularly -CH₂-, >CH-CH₃, -O-;
- n is an integer selected from 1 to 6, particularly 1 to 4;
- m is an integer selected from 0 to 4, particularly 0 to 2;

The structures are not limited to the isomers drawn in formulae I to VIII.

Compounds of formulae I to VIII are known and can be produced according to well known processes.

Polymeric compositions according to the invention comprise the nucleating agent in an amount of from 0,01 to 5% by weight, preferably from 0,02 to 0,5% by weight, more preferably 0,02 to 0,3% by weight based on the polymeric material.

Another object of the invention is a process to enhance the degree of crystallinity, the

crystallization onset temperature and/or mechanical properties of polymeric materials characterized in that

- in a first step at least one nucleating agent selected from the group consisting of compounds of formulae I to VIII optionally mixed with other nucleating agents according to the prior art is added to the unmolten polymeric material and mixed according to known methods forming a mixture,
- in a second step said mixture is melted forming a polymeric composition and
- in a third step said polymeric composition is processed according to known methods

or

ş.

- in a first step at least one nucleating agent selected from the group consisting of compounds of formulae I to VIII optionally mixed with other nucleating agents according to the prior art is added to the molten polymeric material and mixed according to known methods forming a polymeric composition and
- in a second step said polymeric composition is processed according to known methods.

The invention further relates to the use of special imides according to the above-mentioned formulae I to VIII as nucleating agents for polymeric materials.

Especially suitable as nucleating agents are those of formula I in which

R if n=1, is
hydrogen, C_{1.12}alkyl (linear or branched), cyclohexyl, phenyl, biphenyl,
C₇₋₁₂aralkyl, C₇₋₁₂alkaryl;

if n>1

C₁₋₈alkylen (linear or branched), cyclohexylen, phenylen, biphenylen,

C₂₋₁₂aralkylen, C₇₋₁₂alkarylen;

R₁, R₂ are independently selected from hydrogen, C₁₋₁₂alkyl (linear or branched), cyclohexyl, phenyl, biphenyl, C₇₋₁₂aralkyl, C₇₋₁₂alkaryl;

- R₄ hydrogen, C₁₋₈alkyl (linear or branched), cyclohexyl, phenyl, C₇₋₁₂aralkyl, C₇₋₁₂alkaryl;
- R₆ -OH, -NHR₄;

 R_7

n is an integer selected from 1 to 4.

The amount of nucleating agent applied is of from 0,01 to 5%, preferably 0,02 to 0,5%, more preferably 0,02 to 0,3% based on the weight of polymeric material present in the composition.

The use of such nucleating agents leads to enhanced physical properties of polymeric compositions, especially for polyolefins, such properties including improved haze, crystallization temperature and mechanical properties.

Further additives that can be added to a polymeric composition according to the invention include antioxidants, such as sterically hindered phenols, secondary aromatic amines or thioethers, such as described in "Kunststoff-Additive"- Gächter/Müller, 3rd Ed., 1990, pp 42-50, the contents of which is incorporated herein by reference; acid scavengers such as sodium-, magnesium- or calcium stearates or -lactates, hydrotalcite, alkoxylated or hydroxylated amines; U.V. stabilizers such as sterically hindered amine (e.g. N-unsubstituted, N-acyl-substituted or N-alkylated 2,2,6,6-tetra-methyl-piperidinyl compounds, also known as hindered amine light stabilizers HALS); U.V. absorbers (e.g.

2-(2'-hydroxy-phenyl)-benztriazoles, 2-hydroxy-benzophenones, 1,3-bis-(2'-hydroxybenzoyl) benzene-salicylates, cinnamates or oxalic acid diamides); U.V. quenchers (e.g. benzoates or substituted benzoates), antistatic agents, flameproofing agents, softeners, plasticizers, lubricants, guard agents, metal deactivators, complexants, biocides, fungicides, other nucleating agents, fillers, pigments and other impact modifiers.

Polymeric materials whose properties can be enhanced by the nucleating agent according to the invention, include homopolymers, copolymers and polymer blends of:

cellulose acetate; cellulose acetatobutyrate; cellulose acetopropionate; cresol-formaldehyde resins; carboxymethylcellulose; cellulose nitrate; cellulose propionate; casein plastics; casein-formaldehyde resins; cellulose triacetate; ethyl cellulose; epoxy resins; methyl cellulose; melamine-formaldehyde resins; polyamide; polyamidimide; polyacrylonitrile; polybut-1-ene; polybut-2-ene; polybutylacrylate; poly(butylene-terephthalate); poly(ethyleneterephthalate); polycarbonate; poly(chloro-trifluoro-ethylene); poly(diallylphthalate); polyethylene; chlorinated polyethylene; polyether ketone; polyetherimide; polyethyleneoxide; polyethersulfone; polytetrafluoroethylene; phenol- formaldehyde resins; polyimide; polyisobutylene; polyisocyanurate; polymethacrylimide; polymethylmethacrylate; poly(4-methylpent-1-ene); poly(α-methylstyrene); polyoxy-methylene; polyacetal; polypropylene; polyphenylene-ether; polyphenylenesulphide; polyphenylenesulfone; polystyrene; polysulfone; polyurethane; polyvinylacetate; polyvinylalcohol; polyvinylbutyral; chlorinated polyvinylchloride; polyvinylidenefluoride; polyvinylidenechloride; polyvinylfluoride; polyvinylchloride; polyvinylformaldehyd; polyvinylcarbazole; polyvinylpyrrolidone; silicon polymers; saturated polyesters; urea-formaldehyde resins; unsaturated polyesters; polyacrylates; polymethacrylate; polyacrylamide; maleinate resins; phenolic resins; aniline resins; furane resins; carbamide resins: epoxide resins and silicon resins.

Examples of suitable copolymers include:

acrylonitrile/butadiene/acrylate; acrylonitrile/butadiene/styrene; acrylonitrile/methylmeth-acrylate; acrylonitrile/styrene/acrylic ester; acrylonitrile/ethylene-propylene-diene/styrene; acrylonitrile/chlorinated polyethylene/styrene; ethylene/propylene; ethylene/propylene-diene;

ethylene/vinylacetate; ethylene/vinylalcohol; ethylene/tetrafluoroethylene; tetra-fluoroethylene/hexafluoropropylene; methacrylate/butadiene/styrene; melamine/phenol-formaldehyde; polyester blockamide; perfluoro-alkoxy-alkane; styrene/butadiene; styrene/maleic acid anhydride; styrene/a-methylstyrene; vinylchloride/ethylene/methacrylate; vinylchloride/ethylene/vinylacetate; vinylchloride/methylmethacrylate; vinylchloride/ octylacetate; vinylchloride/vinylacetate and vinylchloride/vinylidenechloride.

Preferred polymeric materials whose properties can be improved by the nucleating agents according to the invention are polyolefins such as polyethylene (e.g. high density polyethylene, low density polyethylene, or medium density polyethylene), polybutylene, poly-4-methylpentene and copolymers and mixtures/blends thereof as well as polyesters [e. g. poly-(ethylenterephthalate), poly(butylenterephthalate)], polyamides and polyoxymethylen.

The nucleating agents are especially suitable for the use in polyolefins and their copolymers prepared by using processing catalysts known as Generation II to Generation V catalysts and which may or may have not (preferably have not) been subjected to a catalyst removal step.

By the term "catalyst removal step" used herein is meant a step which comprises, for the purpose of actively removing the catalyst residue contained in the polymerized polyolefins, treating the polyolefins with the compounds which can react with the catalyst residue and inactivate or solubilize the residue, such as e.g. alcohols or water and then removing the inactivated or solubilized catalyst residue by physical means such as filtration, washing, and centrifuging.

Thus, in the case of suspension polymerization, the step of separating the resulting polymer from a dispersion medium such as a solvent or a liquified monomer does not fall under the above-mentioned definition of the catalyst residue-removal step, although the catalyst dissolved in the dispersion medium may be removed at the separation step. The step of adding a small amount of catalyst poisons such as ethers, alcohols, ketones, esters and water to the resulting polymer suspension with a gas such as steam or nitrogen to remove the dispersion medium also does not fall under the above-mentioned definition of the catalyst residue-removal step.

What is meant by Generation I catalysts are titanium halide catalysts and an organo aluminium compound or an organo aluminium halide.

What is meant by Generation II catalysts are Generation I catalysts supported on an organo magnesium compound or based on an organo chromium compound supported on SiO₂.

What is meant by a Generation III catalyst is a Ziegler type complex catalyst supported on a halogen containing magnesium compound.

What is meant by a Generation IV catalyst is a Generation III catalyst with a silane donor.

What is meant by a Generation V catalyst is a bis-indenyl organo titanium compound supported on alumoxane polymers or are bis-cyclopentadienyl-titanium halides activated by aluminium alkyl compounds.

Further generations of highly specific catalysts, especially useful for manufacturing highly stereoregular poly- α -olefins, which are presently under development, belong in the sense of the present invention also to the aforementioned generations of supported catalyst systems. Examples for the microstructure of such highly stereoregular polyolefins are given by syndiotactic polypropylene, isotactic stereoblock polymers, isotactic polypropylene containing steric defects distributed along the polymer chain (so called anisotactic polypropylene) or stereoirregular stereoblock polymers.

Due to the rapid progress in the development of supported newer generation catalyst systems (eg the metallocene catalysts) the commercial significance of these polymers with novel, highly interesting properties increases more and more. However, residues of such further catalyst generations, as long as they contain metals of the 3d, 4d and 5d series of the periodic system supported analogously to the earlier catalyst generations, can also cause disadvantageous properties in the polymer, as long as such residues are not removed physically and are still present in the polymer even in a disactivated form.

These Generations of Catalysts are described in the Twelfth Annual International Conference

on Advances in the Stabilization and Controlled Degradation of Polymers held in Luzern, Switzerland, 21-23 May 1990 in an article on pages 181 to 196 inclusive by Rolf Mülhaupt entitled "New Trends in Polyolefin Catalysts and Influence on Polymer Stability". The contents of this article is incorporated herein by reference and especially Table I on page 184 describing the Generation of Catalysts:

TABLE I Polyolefin Catalyst Evolution

Ge	neration Example	•	ct. % Act. Ti (Ti h atm)	Stereoreg. (%hept.ins)	Process Technology
I .	TiCl ₄ /AIR ₃	40	0.01	45%	removal of cat. residues and atactic PP
	TiCl ₃ /AlEt ₂ Cl	30	0.1	92%	removal of catalyst residues
п	Mg(OEt ₂)/TiCl ₄ /AIR ₃ SiO ₂ /Cp ₂ Cr	40000 _.	HDPE	50%	no removal of cat. residues (mainly HPDE/LLDPE)
ш	Mod.TiCl ₃ cat. MgCl ₂ /TiCl ₄ /AlR ₃ + ester donor	5000 20000	1 10	95% 92%	no purification
īv	MgCl ₂ /TiCl ₄ /AlR ₃ + silane donor	40000	18	99%	no purification no extrusion
v	Bis-indenyl-TiR ₂ on (AlCH ₃ O) ₂	40000	100	99%	novel PPs, narrow MWD

in which R is an organo group; HDPE is high density polyethylene, LLDPE is linear low density polyethyene, Cp is cyclopentadienyl, Et is ethyl, PP is polypropylene, MWD is molecular weight distribution and x is an integer above 2.

Nucleating agents according to the invention may be added to the polymeric material before, during or after the polymerization step and may be added in solid or molten form, in solution preferably as a liquid concentrate or as a solid masterbatch composition containing 5 to 80% (preferably 10 to 50%) by weight of the nucleating agents and 95 to 20% (preferably 90 to 50%) by weight of a solvent or a solid polymeric material which is identical or compatible with the material to be stabilized.

Nucleating agents according to the invention may be incorporated by known methods into the polymeric material. Of importance is dry-blending of the compositions according to the invention with the polymer fluff or coating shaped polymer particles, e.g. polymers spheres, with the nucleating agents in the form of a molten liquid, a solution or suspension/dispersion. Of particular importance is blending of the compounds with thermoplastic polymers in the melt, for example in a melt blender or during the formation of shaped articles, including films, tubes, fibres and foams by extrusion, injection molding, blow molding, roto-molding, spinning or wire coating. The nucleating agents according to the invention are particularly useful for polypropylene and polyethylene, polyester, polyamide and polyoxymethylene articles of every type.

For avoidance of doubt, where a symbol appears more than once in a formula or once in two different formulae, its significances are independent unless indicated to the contrary. Any group capable of being linear or branched is linear or branched unless indicated to the contrary. Further, where a range is given, the figures defining the range are included therein. The abbreviation t-butyl means tertiary butyl, -C(CH₃)₃.

Nucleating agents according to the invention improve the thermal and/or mechanical properties of the polymer material without influencing negatively the processing properties of the said polymeric material. But not all properties may be influenced or improved at the same time. This offers the opportunity to modify exactly the properties needed for a certain application by choosing the right nucleating agent. Furthermore the compounds claimed show advantages concerning polymer compatibility over nucleating agents according to the prior art, e. g. the phthalimides claimed in DE 1 951 632.

The invention will now be illustrated by the following examples.

I. Preparation of the nucleating agents according to the invention

Example I

Synthesis of 1.3-phenylen-N.N-bis-succinimide.

To a stirred solution of 0.1 mol succinic anhydride in 100 ml acetone at 50°C a solution of 0.05 mol 1,3-phenylenediamine in 60 ml acetone is added during 30 min. After 2 h of additional refluxing the solid is filtered off and dried. It is heated with 2 g sodium acetate and 0.21 moles acetic anhydride for 3 h at 70°C. 100 ml water are added, stirred for 1 h, filtered, washed intensively with hot water and dried. Yield: 72%, mp. 203°C.

Example 2

Synthesis of 1.2-phenylen-N.N-bis-succinimide

To a stirred solution of 0.1 mol succinic anhydride in 100 ml acetone at 50°C a solution of 0.05 mol 1,2-phenylenediamine in 60 ml acetone is added during 30 min. After 2 h of additional refluxing the solid is filtered off and dried. It is heated with 2 g sodium acetate and 0.21 moles acetic anhydride for 2 h at 70°C. 150 ml water are added, stirred for 1 h, filtered and recrystallized from hot water and dried. Yield: 32%, mp. >250°C.

Example 3

5

Synthesis of 1.4-phenylene-N,N-bis-succinimide

To a stirred solution of 0.2 mol succinic anhydride in 150 ml acetone a solution of 0.1 mol 1,4-phenylenediamine in 50 ml acetone is added during 30 min. After 2 h of additional refluxing the solvent is stripped in vacuo and 2 g sodium acetate and 0.3 moles acetic anhydride are added. The reaction mixture is stirred for 3 h at 70°C, then the separated colorless crystals are collected

on a Büchner funnel, washed with cold water and dried. The raw product is recrystallized from water. Yield: 42%, mp.: 254 - 256°C.

Example 4

Synthesis of ethylene-N.N-bis-succinimide

To a stirred solution of 0.1 mol succinic anhydride in 50 ml acetone a solution of 0.05 mol ethylenediamine in 50 ml acetone is added during 30 min. After 1 h of refluxing the solid is filtered off and dried. It is heated with 2 g sodium acetate and 0.21 moles acetic anhydride for 2 h at 70°C. 110 ml water are added, stirred for 1 h, filtered and recrystallized from hot water and dried. Yield: 62%, mp. 255°C.

Example 5

Synthesis of (4.4'-diamino-diphenyl-methane)-N.N-bis-succinimide

To a stirred solution of 0.1 mol succinic anhydride in 100 ml acetone a solution of 0.05 mol 4,4'-diamino-diphenyl-methane in 50 ml acetone is added during 30 min. After 1 h of refluxing the solid is filtered off and dried. It is heated with 2 g sodium acetate and 0.21 moles acetic anhydride for 2 h at 70°C. 200 ml water are added, stirred for 1 h, filtered and recrystallized from DMF/water and dried. Yield: 68%, mp. 165°C.

Example 6

Synthesis of ethylene-N.N-bis-(bicyclo[2.2.2]dibenzo-octadien-dicarboximide)

To a stirred solution of 0.1 mol bicyclo[2.2.2]dibenzo-octadien-dicarboxylic anhydride in 200 ml acetone a solution of 0.05 mol ethylendiamine in 50 ml acetone is added during 30 min. After 1 h of reflux 2 g sodium acetate and 0.21 moles acetic anhydride are added and refluxed for 2.5 h. 200 ml water are added, refluxed for 1 h, filtered, recrystallized from dioxane and dried. Yield: 10%, mp. >250°C.

Example 7

Synthesis of ethylene-N.N-bis-(norbon-4-en-dicarboximide)

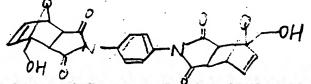
To a stirred solution of 0.1 mol of norbonendicarboxylic anhydride in 100 ml acetone, a solution of 0.05 mols ethylendiamine in 100 ml acetone is slowly added. After refluxing the mixture for 2 h, it is cooled, filtered and dried. This substance is stirred for 2 h at 60°C with a mixture of 2. g sodium acetate and 10 ml acetic anhydride. 50 ml water are added, stirred for 1 h, filtered and dried. The raw product is recrystallized from toluene. Yield: 43%, mp. 244 - 248°C.

Example 8

Synthesis of phenyl-N-bis-(1-(hydroxy-methyl)-7-oxo-norbon-4-en-dicarboximide)

To a stirred solution of 0.1 mol maleic anhydride in 150 ml acetone a solution of 0.1 mol aniline in 50 ml acetone is added during 30 min. After 2 h of reflux 2 g sodium acetate and 0.21 moles acetic anhydride are added and refluxed for 2.5 h. 200 ml water are added, refluxed for 1 h, filtered and recrystallized from cyclohexane and dried. This substance is reacted with 0.075 mols furfurylalcohol in 100 ml toluene at 70°C for 12 h. The product is filtered off and recrystallized from ethyl acetate. Yield: 58%, mp. 154°C.

Example 9



Synthesis of 1.4-phenylene-N.N-bis-(1-(hydroxy-methyl)-7-oxo-norbon-4-en-dicarboximide)

To a stirred solution of 0.2 mol maleic anhydride in 150 ml acetone a solution of 0.1 mol 1,4-phenylenediamine in 50 ml acetone is added during 30 min. After 1 h of reflux 2 g sodium acetate and 0.21 moles acetic anhydride are added and refluxed for 5 h. 200 ml water are added, refluxed for 1 h., filtered after cooling and recrystallized from DMF and dried. This substance is reacted with 0.1 mol furfurylalcohol in 100 ml toluene at 70°C for 10 h. The product is filtered off and washed with methanol. Yield: 27%, mp. >250°C.

Example 10

D-NOC WH

Synthesis of trimellitic acid imide-anilide

To 0,1 mol of the methyl ester of trimellitic acid imide 0,1 mol aniline and 100 ml decalin is added. The mixture is heated until the theoretical amount of methanol is distilled off (12 h.). After cooling the precipitated product is filtered off and dried. For further purification the raw product is recrystallized from dimethylacetamide. Yield: 86%, mp. >250°C.

Example 11 = Comparative example according to the prior art

Synthesis of 1.3-phenylen-N.N-bis-phthalimide

To a stirred refluxing solution of 0.2 mol phthalic anhydride in 200 ml acetic acid a solution of 0.1 mol 1,3-phenylenediamine in 100 ml acetic acid is added during 30 min. After 2 h of additional stirring the off white precipitate is collected on a Büchner funnel, washed neutral with water, acetone and dried. Yield: 62%, mp.: >250°C.

II. Production of the polymeric compositions according to the invention

Examples 12 - 17

Polymeric compositions containing

100.0 parts	3 rd generation random polypropylene copolymer (MFI 7)
0.05 parts	Irganox [®] 1010
0.1 parts	Sandostab® P-EPQ
0.1 parts	calcium stearate
0.1 parts	nucleating agent according to table 1 below

are prepared by dry blending the components and extruding this mixture at 210°C on a Händle single screw extruder. The resulting pellets are then injection molded on an Arburg injection

molding machine at 245°C to form plaques of $100 \times 100 \times 1 \text{ mm}^3$.

Examples 18 to 22

Polymeric compositions containing

100.0 parts	3 rd generation random polypropylene copolymer (M	FI 7)
0.05 parts	Irganox [®] 1010	
0.1 parts	Sandostab® P-EPQ	
0.1 parts	calcium stearate	
0.3 parts	nucleating agent according to table 1 below	•

are prepared according to the procedure given in the examples 12 to 17.

Examples 23 to 26

Polymeric compositions containing

100.0 parts	3 rd generation random polyp	ropylene copolyme	r (MFI 6)
0.05 parts	Irganox [©] 1010		
0.1 parts	Sandostab® P-EPQ		• =
0.1 parts	calcium stearate		
0.1 parts	nucleating agent according t	o table 1 below	

are prepared according to the procedure given in the examples 12 to 17.

Examples 27 to 29

Polymeric compositions containing

100.0 parts	3rd generation random polypr	opylene copo	olymer ((MFI	6)
0.05 parts	Irganox® 1010	•	•		

0.1 parts Sandostab® P-EPQ

0.1 parts calcium stearate

0.3 parts nucleating agent according to table 1 below

are prepared according to the procedure given in the examples 12 to 17.

The following properties are determined:

- the melt flow index of the granules of the polymeric composition after the first extrusion according to ASTM D 1238-70/L
- the yellowness index of the injection molded plaques according to ASTM D 1925-70 (reflection)
- the haze values of the injection molded plaques according to ASTM D 1003/70
- the crystallisation onset temperature (COT). For this purpose samples of about 3 mg are punched out of the middle of the plaques and the COT is determined with a Mettler DSC 20 (200°C to 50°C, -10°C min⁻¹).
 - The textile strength (TS) according to DIN 53 455:

From the injection molded plaques samples are cut perpendicular to the injection direction. The values for TS given in the tables below are the averages of 5 measurements.

All test results are summarized in the tables 1 and 2.

Table 1

Results of polymeric compositions according to the invention and of a polymeric composition without a nucleating agent

Example No.	Nucleating agent of	Amount of	Haze	COT	TS
	Symple	nucleating agent [parts]	[%]	ູເລີ	N/31
12	-	0.1	300		(IVIIIIII)
		1.0	33	601	26.1
	2	0.1	27	011	27.6
14		0.1	. 59	122	27.2
51	. 4	0.1	27	601	25.8
91	9	0.1	43	611	25.5
17		•	27	108	24.9
8 1		0.3	55	109	25.5
61	2	0.3	40	011	24.6
20	8	0.3	79	118	27.4
21	4	0.3	32	109	26.5
22	6	0.3	57	801	24.6

Table 2

Results of polymeric compositions according to the invention and of a polymeric composition without a nucleating agent

Example No.	Nucleating agent of Example	Amount of	Haze	COT	TS
		[parts]	[%]	[2]	[N/mm²]
23	L	0.1	23	104	28.9
24		0.1	22	106	32.4
25	6	0.1	24	105	30.9
26			26	105	29.4
27	7	0.3	25	<u> 104</u>	29.6
28	· ·	0.3	30	104	32.9
29	6	0.3	55	110	30.0